

Beryllium Waste Transuranic Inventory in the Subsurface Disposal Area, Operable Unit 7-13/14

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ABSTRACT

Between 1970 and 1993, irradiated reactor beryllium reflector waste from the Advanced Test Reactor, Engineering Test Reactor, and the Materials Test Reactor was buried in the Subsurface Disposal Area. The radionuclide inventories associated with these beryllium blocks were based on characterization methods and measured data.

This report documents a study performed to apply new characterization data to inventory estimates associated with irradiated beryllium disposed of in the Subsurface Disposal Area from the Advanced Test Reactor, Engineering Test Reactor, and the Materials Test Reactor. Chemical assay data obtained from irradiated beryllium currently stored in the Advanced Test Reactor canal were used to validate the calculated inventories associated with the disposed-of beryllium waste at the Subsurface Disposal Area. The computational models that were used to perform this study, along with the data on which they were based, are discussed in some detail. Also included is a discussion of the irradiated beryllium not yet disposed of that will eventually require disposal.

EXECUTIVE SUMMARY

Beryllium is used as a neutron reflector in many research reactors around the world. At the Idaho National Engineering and Environmental Laboratory Test Reactor Area, beryllium has been used in three reactors: the Advanced Test Reactor (ATR), the Engineering Test Reactor (ETR), and the Materials Test Reactor (MTR). Between 1970 and 1993, beryllium reflectors and other components irradiated in those three facilities were buried as radioactive low-level waste in the Subsurface Disposal Area (SDA) at the Radioactive Waste Management Complex of the Idaho National Engineering and Environmental Laboratory. New information concerning radionuclide concentrations in irradiated beryllium, now being stored in the ATR canal, indicates that previous characterizations of irradiated beryllium are incorrect. These new data have reduced the uncertainty in the beryllium characterization data compared to earlier characterization data.

Though a number of radioisotopes were modeled and documented in this study, the analysis focused on C-14 and several transuranic waste (TRU) isotopes identified as potential risk drivers for Operable Unit 7-13/14 (Becker et al. 1998). The conclusions derived from this study follow:

- The C-14 inventory in the buried MTR, ETR, and ATR beryllium is approximately 92.4 Ci. The estimated TRU inventories in buried beryllium are small compared to the total TRU inventories in the SDA.
- Before this study, it was unknown whether uranium was present in sufficient concentrations in the ATR beryllium to produce a waste with a significant TRU inventory. This study not only verifies that TRU concentrations are present in the ATR beryllium, but that these concentrations exceed 100 nCi/g for TRU. Also, this study concludes that the beryllium reflectors from the first core loadings of the MTR and ETR (material that was disposed of in the SDA) exceed the 100-nCi/g limit for TRU.
- Approximately 3.6 Ci of TRU isotopes are attributable to the disposal of MTR, ETR, and ATR beryllium.
- Computer modeling for characterization of the ATR beryllium blocks and outer shim control cylinders (OSCCs) was validated by comparing model concentrations to actual measured concentrations for several isotopes at two sample point locations on ATR Block 010R. This study is the first case in which significant measured data have been obtained on the ATR beryllium core components and then compared against computer code results. The results of these comparisons have shown that the best-estimate computer code results are generally within a factor of 2 for most measured data. The data presented in this report represent the best-estimate characterization currently available for approximately 40 radionuclides considered in this analysis.
- The calculated Pu-239 and TRU values using both the segmented model and the single-block model (computed at two different sites on the block) are in very good agreement (usually within a factor of 2 or better) and are in agreement with the sample assay measurements taken from the two sites modeled. This information provides validation of the computer models and the basis for the

segmented and single-block computer models to accurately determine the total block inventory of TRU isotopes and other radionuclides of interest.

- Based on the segmented three-dimensional model calculation, more than 96% of the beryllium block mass exceeds 100 nCi/g. In addition, nearly 50% of the block mass exceeds 400 nCi/g.
- The calculated TRU total inventory per gram for Block 010R, based on the (3-D) segmented computer model, was 277.08 nCi. The corresponding single-block (1-D) Oak Ridge Isotope GENERation and Depletion Code Version 2 model calculated a TRU total block inventory per gram of metal to be 491.23 nCi. (See Tables 6-4 or 7-12. Note: in Table 7-12, the 010R inventory is the same as that shown for Block 015L.) The accuracy of both computer code models is about a factor of two; however, based on reasons explained in Section 6.7, the (1-D) Oak Ridge Isotope GENERation and Depletion Code Version 2 results are considered to be best estimate.
- The TRU concentration is a dynamic quantity that can be accurately determined from computer analysis. The primary variables that influence the TRU inventory are irradiation time, initial uranium inventory, neutron flux, and decay time. During irradiation, the TRU inventory tends to build up until a maximum value is reached before the TRU isotopes begin to burn out and decrease the total TRU concentration. However, following irradiation, the TRU isotopes significantly increase in concentration because of the beta decay of the non-TRU isotope Pu-241 into the TRU isotope Am-241.
- The segmented model provides insight into the three-dimensional distribution of important isotopes within a typical ATR beryllium block. This distribution includes TRU isotopes, C-14, and H-3. The segmented three-dimensional model also is a powerful tool for determining alternative or representative sampling locations for future sampling.
- The elemental assay data presented in this report represent the best-estimate chemical impurity information that is known for beryllium reflector material used at the Idaho National Engineering and Environmental Laboratory and in the industry. In addition, best-estimate reactor operating conditions were assumed in the calculations for the MTR, ETR, and ATR beryllium components.
- Before this investigation, it was not known how many Core 2 blocks had been disposed of. The current investigation indicates that two of the Core 2 blocks still reside in the ATR canal and that six Core 2 blocks were disposed of on or about June 1, 1977, in the SDA. Though it was originally known that nine OSCCs had been disposed of in the SDA, whether all of these shim cylinders came from the Cores 1 and 2 irradiation was not known. This study has confirmed that the OSCCs came from Cores 1 and 2, but which core positions these OSCCs occupied still are unknown. The serial numbers for the buried OSCCs and many of the reflector block serial numbers are not known.

RECOMMENDATIONS

The following recommendations were identified during the preparation of this report. Some are directly applicable to the beryllium materials disposed of in the Subsurface Disposal Area (SDA). Others are related to beryllium materials stored in the Advanced Test Reactor, Materials Test Reactor (MTR), and Engineering Test Reactor (ETR).

Recommendation for Beryllium Materials Disposed of in the Subsurface Disposal Area

According to information presented in this report, the beryllium reflectors from MTR, ETR, and Advanced Test Reactor that were disposed of in the SDA are now considered to be a transuranic waste; however, information from R. D. Gibby (EDF-2101) indicates that the beryllium metal is a Resource Conservation and Recovery Act nonhazardous material. Because of a higher-than-expected corrosion rate of beryllium metal in the SDA and the high inventory of C-14 contained within this material (relative to other waste in the SDA that contain C-14), it is recommended that some remedial efforts be undertaken to reduce the release rate of C-14 from the disposed-of beryllium at the SDA.

Recommendations for Beryllium Materials Remaining in the Advanced Test Reactor, Materials Test Reactor, and Engineering Test Reactor

Though the MTR and ETR characterization data in this report apply specifically to beryllium disposed of from these reactors, with slight revision it could be applied to beryllium reflector materials and other core components remaining in the ETR and MTR reactors to support future waste disposal for facility deactivation, decontamination, and decommissioning. The *Characterization of the Engineering Test Reactor Facility* (Kaiser et al. 1982) and *Characterization of the Materials Testing Reactor* (Rolfe and Wills 1984) reports should be revised to reflect information provided in this report.

All beryllium reflector material currently stored in the Advanced Test Reactor canal and beryllium material to be generated in the future should be modeled to estimate the total radiological inventory to support future waste path planning. The modeling should be expanded also to estimate the radiological inventory that will result from continued operation of the Advanced Test Reactor Critical reactor.

Finally, laboratory analyses of additional samples from irradiated beryllium currently stored at the Idaho National Engineering and Environmental Laboratory and enhanced computer modeling efforts can be used to improve the characterization of future beryllium reflector materials.

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ACRONYMS

ANL-W	Argonne National Laboratory-West
ATR	Advanced Test Reactor
ATRC	Advanced Test Reactor Critical (facility)
BOL	beginning of life
CFR	<i>Code of Federal Regulations</i>
CIC	core internal changeout
D&D&D	deactivation, decontamination, and decommissioning
EOI	end of irradiation
EOL	end of life
ETR	Engineering Test Reactor
ETRC	Engineering Test Reactor Critical (facility)
INEEL	Idaho National Engineering and Environmental Laboratory
KBI	Kawecki Berylco Industries
LLW	low-level waste
MCNP4B	Monte Carlo N-Particle Version 4B
MOCUP	MCNPB4-ORIGEN2 Coupled Utility Program
MTR	Materials Test Reactor
ORIGEN2	Oak Ridge Isotope GENeration and Depletion Code Version 2
OSCC	outer shim control cylinder
OU	operable unit
RI/FS	remedial investigation/feasibility study
RWMIS	Radioactive Waste Management Information System
RWMC	Radioactive Waste Management Complex
SDA	Subsurface Disposal Area
SVR	soil vault row

TRA	Test Reactor Area
TRU	transuranic waste
wppm	weight parts per million

Beryllium Waste Transuranic Inventory in the Subsurface Disposal Area, Operable Unit 7-13/14

1. INTRODUCTION

Beryllium is used as a neutron reflector in many research reactors around the world. At the Idaho National Engineering and Environmental Laboratory (INEEL) Test Reactor Area (TRA), beryllium has been used in three reactors: the Advanced Test Reactor (ATR), the Engineering Test Reactor (ETR), and the Materials Test Reactor (MTR). Between 1970 and 1993, beryllium reflectors and other components irradiated in those three facilities were buried as radioactive low-level waste (LLW) in the Subsurface Disposal Area (SDA) at the Radioactive Waste Management Complex (RWMC) of the INEEL. New data concerning radionuclide concentrations in irradiated beryllium, now being stored in the ATR canal, mean that previous characterizations of irradiated beryllium were incorrect. These new data have reduced the uncertainty in the beryllium characterization data compared to earlier characterization data.

1.1 Purpose

The purpose of this report is to provide estimates of radionuclide inventories associated with beryllium blocks disposed of in the SDA. The inventory will be used to support the development of the comprehensive remedial investigation/feasibility study (RI/FS) for Waste Area Group 7. The RI/FS will ultimately provide basis for the remedial decision-making under the Federal Facility Agreement and Consent Order (DOE-ID 1991).

1.2 Scope

Newly developed beryllium characterization data and methods support disposal planning for beryllium reflector waste stored in the ATR canal. These data and methods were used to revise and improve the accuracy of the radiological source term for irradiated beryllium waste from the ATR, ETR, and MTR reactors. That waste has been disposed of by burial in the SDA. The new information has been incorporated into this report to provide improved data for characterizing the radiological properties of beryllium that have previously been disposed of and for possible future disposal of irradiated beryllium waste. The computational models central to this characterization, their inputs and outputs and the measurements on which they were based, are discussed in some detail. Also included is a discussion of the irradiated beryllium not yet disposed of that will eventually require disposal. This material is in the ATR canal; in the ATR, MTR, and ETR reactors; and waiting or planned for placement in the ATR.

1.3 Background

The data in this report support the Operable Unit 7-13/14 comprehensive RI/FS required under the Federal Facility Agreement and Consent Order (DOE-ID 1991) and the Comprehensive Environmental Response, Compensation, and Liability Act (42 USC § 9601 et seq., 1980). For management purposes, under the Federal Facility Agreement and Consent Order the INEEL was divided into 10 waste area groups. Waste Area Group 7, comprising the RWMC, is located in the southwest quadrant of the INEEL. Operable Unit (OU) 7-13/14 is the comprehensive RI/FS for Waste Area Group 7. The INEEL, the RWMC, and other facilities are shown in Figure 1-1. A map of the RWMC showing the SDA is shown in Figure 1-2.

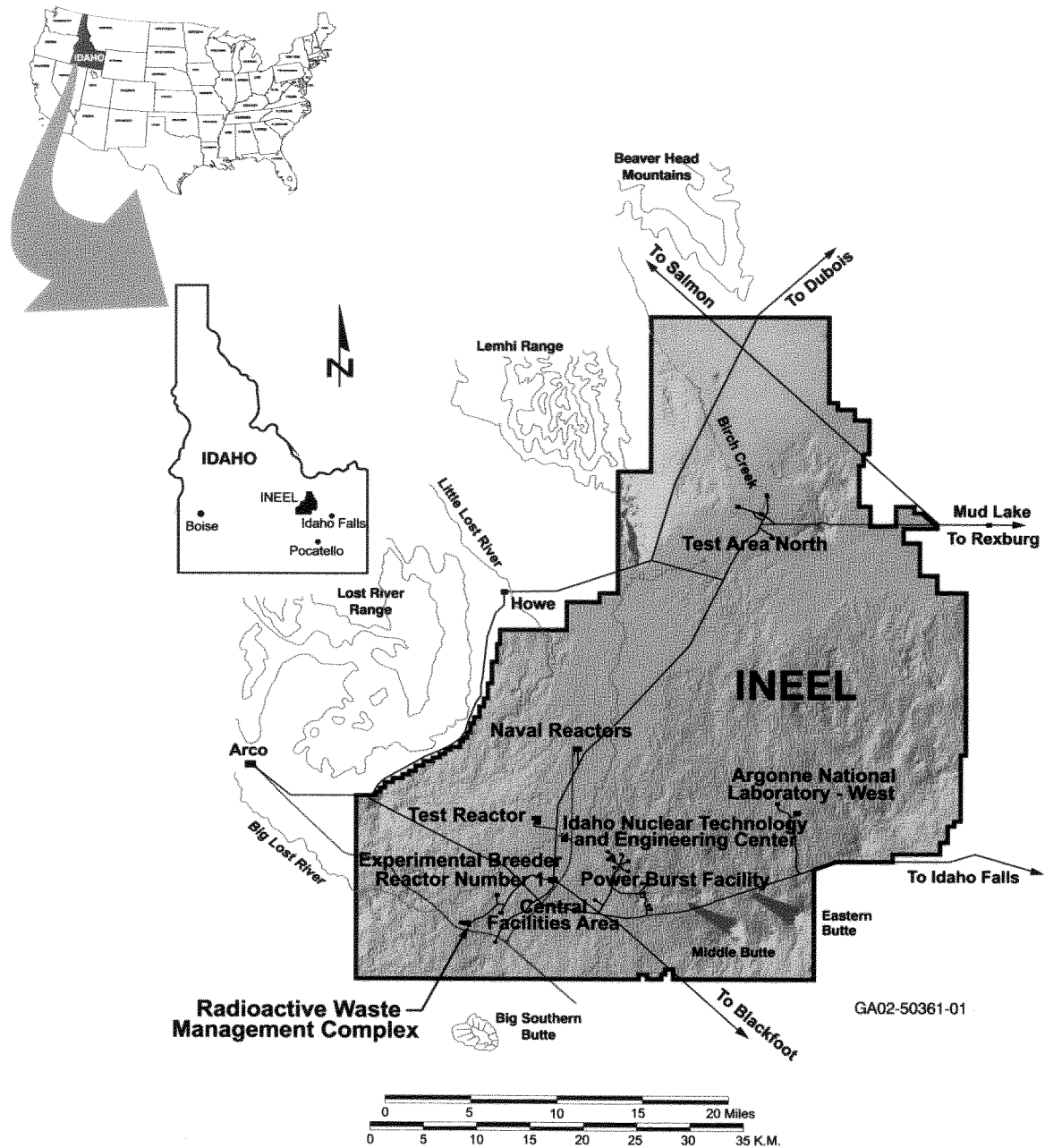


Figure 1-1. Relief map of the Idaho National Engineering and Environmental Laboratory showing the location of the Radioactive Waste Management Complex and other facilities.

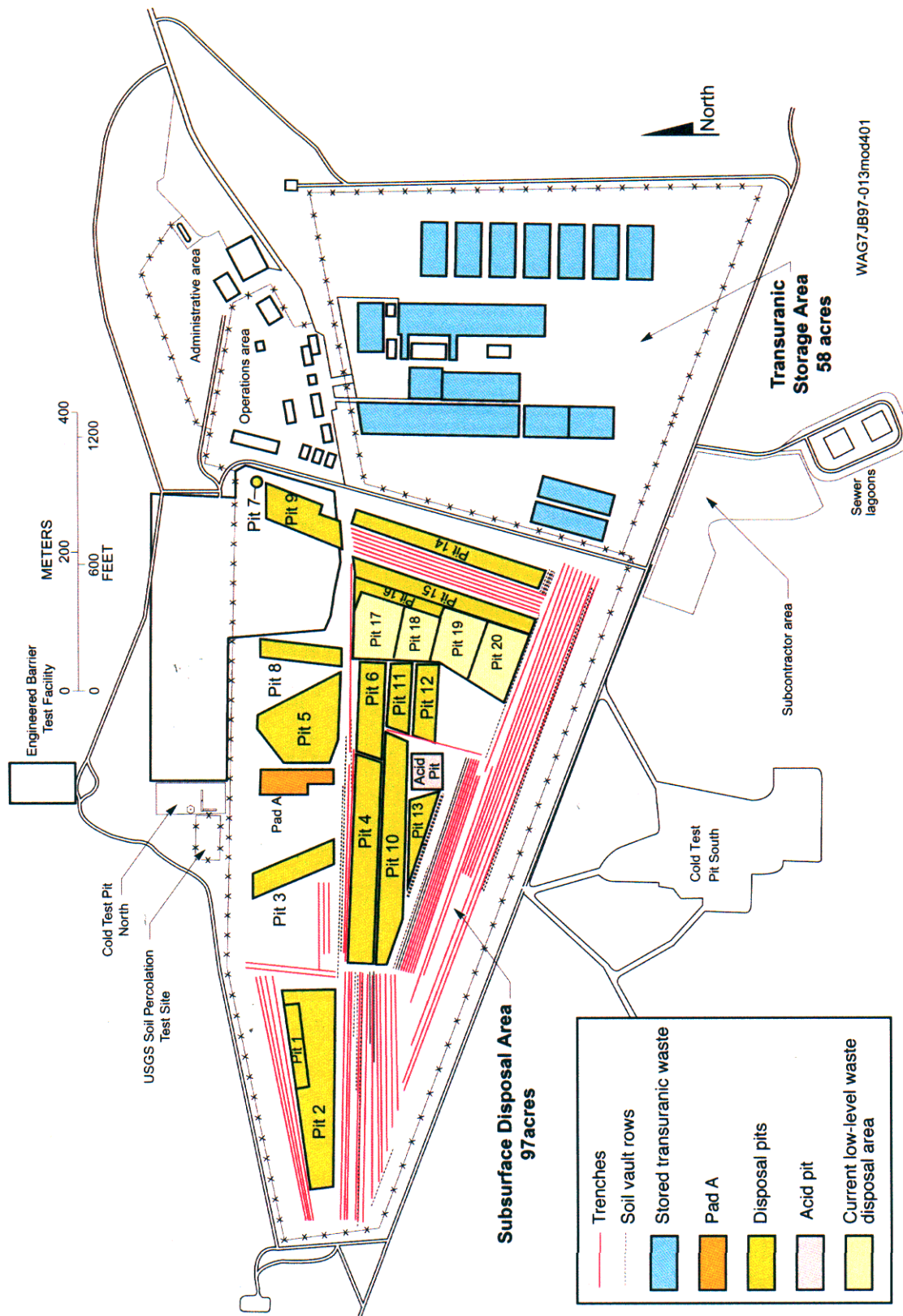


Figure 1-2. Map of the Radioactive Waste Management Complex showing location of the Subsurface Disposal Area.

The boundary of Waste Area Group 7 is defined as the RWMC fence. The SDA is a fenced portion within the RWMC, which includes numerous pits, trenches, and soil vaults where transuranic (TRU) waste and LLW have been buried since 1952, though disposal of hazardous material in the SDA ceased in 1983. Since 1983, only LLW that met the requirements of the INEEL *Reusable Property, Recyclable Materials, and Waste Acceptance Criteria* (DOE-ID 2002) has been disposed of in the SDA active LLW disposal pit.

The MTR, the ETR, and the currently operating ATR at the INEEL use beryllium reflectors. The ATR reflector consists of a set of eight beryllium blocks and 16 outer shim control cylinders (OSCCs). For the ETR, the reflector is essentially four slabs of beryllium that surround the core, while for the MTR, the reflector is much more complex. For the ATR, the reflector and OSCCs must be replaced periodically because of swelling, generally at 8- to 10-year intervals.

Four ATR core internal changeouts (CICs) have been completed to date and the fifth is scheduled for 2004. From 1970 through 1993, some of the beryllium removed from the cores of the reactors at TRA was disposed of as LLW in the SDA. Beryllium reactor waste disposed of includes 20 beryllium blocks from ATR Cores 1, 2, and 3, nine OSCCs from ATR Cores 1 and 2, and one beryllium reflector assembly each from the MTR and ETR. This was verified by evaluation of current storage and disposal documentation.

The ETR Critical (ETRC) facility was a low-power reactor that supported ETR operations. It had a beryllium reflector of nominally the same dimensions as the ETR. The ETRC beryllium reflector appears to have been removed from the reactor and disposed of—most likely in the SDA. However, actual disposal records were not found to support this conclusion. The ETRC beryllium reflector has been included in this study to provide completeness. Because of the low power level of the ETRC, its beryllium reflector, if disposed of in the SDA, contributes negligibly to the C-14 inventory and to the TRU totals.

The majority of irradiated beryllium reflector waste was disposed of in the SDA in three major disposal campaigns: 1976, 1977, and 1993. A total of 4,742 kg (10,454 lb) of beryllium was disposed of from the ATR, ETR, and MTR.

Previous characterizations of beryllium buried at the SDA relied on modeling alone to develop the inventories of isotopes of concern, and it was believed that certain key nuclides like C-14 were overestimated in the inventory. This led to a cessation in the disposal of the blocks. In an effort to reduce the conservatism in the inventory estimates, samples were taken from beryllium blocks irradiated in the ATR but still in the ATR canal. The sampling showed that the initial models were indeed conservative with regard to C-14 but substantially underestimated the Nb-94 concentrations because of incorrect information on Nb-93 impurity. They also revealed uranium impurity in the blocks that had not been identified before the sampling. This impurity is important because, under the neutron flux in a reactor, the uranium is transmuted to transuranic isotopes. Further investigation has shown that the blocks contain a high enough transuranic content to be classified as TRU. This classification is important for the OU 7-13/14 RI/FS because it has implications for possible remedial action regarding the blocks buried at the SDA.

Characterization improvements have been made for the specific isotopes C-14, Nb-94, and alpha-emitting TRU. These data were obtained through comparison of results from a new three-dimensional cross-section computer model of the ATR with measurements on samples taken from ATR reflector blocks, refinement of beginning-of-life chemical impurity estimates, and improved specificity of neutron exposure. The processes and programs used to ensure that the generated data will be suitable for the intended use are described in this report. The scope of this work also includes the development of a detailed radiological computer model for producing characterization data for the MTR and ETR reactors.

1.4 Findings

Based on the disposal dates, measurements, and calculations reported here, a total of 92.4 Ci of C-14 was disposed of in the SDA in beryllium reflector material from MTR, ETR, and ATR. Nb-94 concentrations were found to be much higher than previously thought because of higher concentrations of the Nb-93 impurity precursor. Nb-94 inventories were explicitly calculated for the MTR, ETR, and ATR beryllium reflectors. These calculations show an Nb-94 inventory of 0.0773 Ci in MTR, 0.0482 Ci in ETR, and 0.0835 Ci in the ATR beryllium. Even though the C-14 inventory is lower than was previously thought, the presence of higher Nb-94 concentrations indicates that the value calculated using the sum-of-fractions rule will exceed one; that is, the irradiated beryllium will exceed the Class C criteria for disposal in 10 *Code of Federal Regulations* (CFR) 61 (2002). Other isotopes in the beryllium material buried at the SDA are summarized in Table 1-1 and are reported in Section 7.

Irradiated beryllium buried at the SDA qualifies as TRU. In scaling the results of calculations and measurements on ATR beryllium to reflectors from the ETR and MTR buried at the SDA, analysis showed that reflectors from both those reactors were transuranic and at higher TRU isotope concentrations than the beryllium from the ATR reflectors. As indicated in Table 1-1, approximately 3 Ci of TRU isotopes were in the reflectors from the ETR and MTR. Some increase in activity levels is a consequence of the lower neutron flux in ETR and MTR beryllium that burned away fewer of the transuranic isotopes produced while in the core. Also, the longer time the ETR and MTR reflectors have been out of the reactor, as compared with ATR beryllium, has allowed greater build-up of transuranic isotopes from their precursors.

Improved computer models and cross sections give good agreement between predictions and measurements. This means that these tools developed for characterizing ATR beryllium should give good predictions for beryllium from the ETR and the MTR. These models and results are discussed in Section 6.

Transuranic waste isotope concentrations in irradiated beryllium change with time. Because of decay of non-TRU precursors, the TRU activity generally increases for several tens of years after the beryllium has been removed from the reactor. Burnout of some isotopes means that the greatest TRU activity will not necessarily be at the location where neutron fluence is the highest.

Table 1-1. Summary of the Advanced Test Reactor, Engineering Test Reactor, and Materials Test Reactor irradiated beryllium reflector waste disposed of in the Subsurface Disposal Area.

Reactor and Beryllium Waste Disposed of by Serial Number or Core Position	Initial Irradiated Date	Final Irradiated Date	Core	Reactor Position	Total Core (MWd)	Be Metal Mass (g)	Metal Volume (m ³)	Disposal Date	Disposal Location	Total C-14 (Ci)	C-14 Concentration (Ci/m ³)	TRU Concentration (nCi/g)	Total TRU per Item (Ci)
Materials Test Reactor	3/31/52	7/3/69	1	N/A	177,887*	~2,000,000*	~1.08	1977	Trench 58 3+10-20 to 3+40-50	29.2	27.00	1,387.00	2.770
Engineering Test Reactor	10/15/57	3/1/70**	1	N/A	~380,000**	~624,000**	~0.337	1970	Trench 52, 4+175, 4+70, 4+85, 4+50	21.7	64.40	257.00	0.160
Advanced Test Reactor													
Block NW-L	2/1/68	9/9/72	1	NW	28,894	81,420	0.044	1976	Trench 58 200+05-15	0.9997	22.72	321.97	0.026
NW-R	2/1/68	9/9/72	1	NW	28,894	81,420	0.044	1976	Trench 58 200+05-15	0.9997	22.72	321.97	0.026
NE-L	2/1/68	9/9/72	1	NE	27,960	81,420	0.044	1976	Trench 58 200+25-35	0.9679	22.00	321.12	0.026
NE-R	2/1/68	9/9/72	1	NE	27,960	81,420	0.044	1976	Trench 58 200+25-35	0.9679	22.00	321.12	0.026
SW-L	2/1/68	9/9/72	1	SW	27,978	81,420	0.044	1976	Trench 58 200+25-35	0.9683	22.01	321.13	0.026
SW-R	2/1/68	9/9/72	1	SW	27,978	81,420	0.044	1976	Trench 58 200+25-35	0.9683	22.01	321.13	0.026
SE-L	2/1/68	9/9/72	1	SE	28,017	81,420	0.044	1976	Trench 58 200+25-35	0.9696	22.04	321.23	0.026
SE-R	2/1/68	9/9/72	1	SE	28,017	81,420	0.044	1976	Trench 58 200+25-35	0.9696	22.04	321.23	0.026
Block NE-L	2/5/73	4/11/77	2	NE	23,625	81,420	0.044	1977	Trench 58 3+10-20	0.8189	18.61	191.90	0.016
NE-R	2/5/73	4/11/77	2	NE	23,625	81,420	0.044	1977	Trench 58 3+10-20	0.8189	18.61	191.90	0.016
SW-L	2/5/73	4/11/77	2	SW	36,285	81,420	0.044	1977	Trench 58 3+10-20	1.2540	28.50	197.52	0.016
SW-R	2/5/73	4/11/77	2	SW	36,285	81,420	0.044	1977	Trench 58 3+10-20	1.2540	28.50	197.52	0.016
SE-L	2/5/73	4/11/77	2	SE	24,357	81,420	0.044	1977	Trench 58 3+10-20	0.8441	19.18	193.52	0.016
SE-R	2/5/73	4/11/77	2	SE	24,357	81,420	0.044	1977	Trench 58 3+40-50	0.8441	19.18	193.52	0.016
Block 018L	8/9/77	2/2/86	3	NW	53,924	81,420	0.044	1993	SVR 20 0+315	1.8530	42.11	387.15	0.032
013R	8/9/77	2/2/86	3	NW	53,924	81,420	0.044	1993	SVR 20 0+315	1.8530	42.11	387.15	0.032
015L	8/9/77	2/2/86	3	NE	47,259	81,420	0.044	1993	SVR 20 0+315	1.6270	36.98	406.25	0.033
019L	8/9/77	2/2/86	3	SW	60,205	81,420	0.044	1993	SVR 20 0+315	2.0660	46.95	370.37	0.030
014R	8/9/77	2/2/86	3	SW	60,205	81,420	0.044	1993	SVR 20 0+315	2.0660	46.95	370.37	0.030
011R	8/9/77	2/2/86	3	SE	72,984	81,420	0.044	1993	SVR 20 0+315	2.4970	56.75	340.98	0.028
Nine outer shim control cylinders	2/5/77	4/11/77	1&2	unknown	165,928 ^a	489,881	0.2648 ^a	1987	SVR 17 0+10, 0+18, 1+00, 1+56	15.9100	60.08	297.01	0.145
Total						4,742,281	2.562			92.4170			3.564
L = left NE = northeast													
MW = megawatt NW = northwest													
MWd = megawatt-day R = right													
SVR = soil vault row SW = southwest													
N/A = not applicable SE = southeast													
TRU = transuramic waste													
a. Total for nine outer shim control cylinders													

* 177.887 MWd is reported by the *MTR Progress Report Cycle No. 295* (Ford et al. 1969). An alternate value of 179,329 MWd (71,322 + 108,000) can be determined from data shown on pages 10 and 11 of *Characterization of the Materials Testing Reactor* (Rolfe and Wills 1984). Rolfe and Wills (1984) also report that the total operating history of MTR (to August 21, 1970) was about 180,000 MWd. The MTR beryllium mass is based on dimensional data (IAEA 1959) and a beryllium density of 1.85 g/cm³. The uncertainty in the MTR beryllium mass is approximately ±14%.

** The final irradiation date for ETR before removal of the original beryllium reflector is estimated to be March 1, 1970. As noted on page 35 of *Characterization of the Engineering Test Reactor Facility—September 1982* (Kaiser et al. 1982), the original beryllium reflector was replaced sometime in March 1970 (but the exact day of the month is not reported). The total number of MWd of ETR operation to March 1970 has not been identified. However, the number of MWd of ETR operation to January 11, 1970, is reported as 374,498 MWd on page 8 of *ETR Operations Branch Progress Report for Cycle No. 105 November 7, 1969—January 11, 1970* (Smith et al. 1970). Also, Kaiser et al. (1982) report 487,728 MWd of ETR operation through 1972. The estimated value of 380,000 MWd is based on an extrapolation of the January 11, 1970, value. The uncertainty associated with this value is ±2%. The ETR beryllium mass is calculated from dimensional data (IAEA 1964; Tobias 1969) and assumes a beryllium density of 1.85 g/cm³. The uncertainty in the ETR beryllium (reflector) mass is estimated to be about ±1%.

1.5 Overview of Methodology and Assumptions

The characterization methodology is based on calculating the radionuclide concentrations in irradiated beryllium reflector materials (blocks and OSCCs) from the ATR. This work is based on new characterization data and improved computer modeling performed for the beryllium reflectors stored in the ATR canal. The computer modeling focused on one of the ATR beryllium blocks, Block 010R, from Core 3 that is currently stored in the canal. The ATR beryllium block and OSCC characterizations and block validation results are then used to scale characterization data for the MTR and ETR beryllium.

The logical flow of characterization activities is presented in Figure 1-3. It identifies the separate tasks and captures the complex process logic that has evolved over the course of the characterization efforts, specific to irradiated beryllium generated by and removed from the ATR, MTR, and ETR. Characterization efforts are focused on ATR beryllium rather than beryllium from MTR and ETR for the following reasons: (a) more complete operational records are available, (b) stored blocks are available for sampling, and (c) a path to disposal must be identified for stored ATR beryllium blocks.

The central focus of the ATR beryllium characterization is the Monte Carlo N-Particle Version 4B (MCNP4B) and Oak Ridge Isotope GENERation and Depletion Code Version 2 (ORIGEN2) models used in the computer calculations. Significant effort has been expended to validate these models against measured assay data to ensure adequacy, accuracy, and confidence. Use of these models, and the radionuclide inventories produced by these models, are the bases for the activation product concentrations for C-14, TRU, and other reported radioisotope estimates in the irradiated beryllium buried in the SDA.

To characterize the MTR, ETR, and ATR beryllium, the authors assumed that the ATR beryllium Block 010R is similar in both physical and radiological properties to the beryllium materials in all three reactor cores. This is a very logical assumption because the beryllium blocks in the SDA are from ATR Cores 1, 2, and 3, and the beryllium blocks remaining in the canal are from Cores 2, 3, and 4. For the MTR and ETR beryllium, a similar assumption is not quite as strong. The beryllium supplier is not known, and no measured data exist for the MTR and the ETR beryllium. The manufacturer of the MTR and ETR beryllium is assumed to be the same supplier as the early ATR beryllium material because the operation time period of the MTR and ETR overlapped, to some degree, that of the ATR beryllium procurements. The assumption is that beryllium procured during concurrent time periods would have had similar characteristics. Sections 3 and 4 provide a more in-depth development of the relationships between the MTR, ETR, and ATR beryllium reflector material.

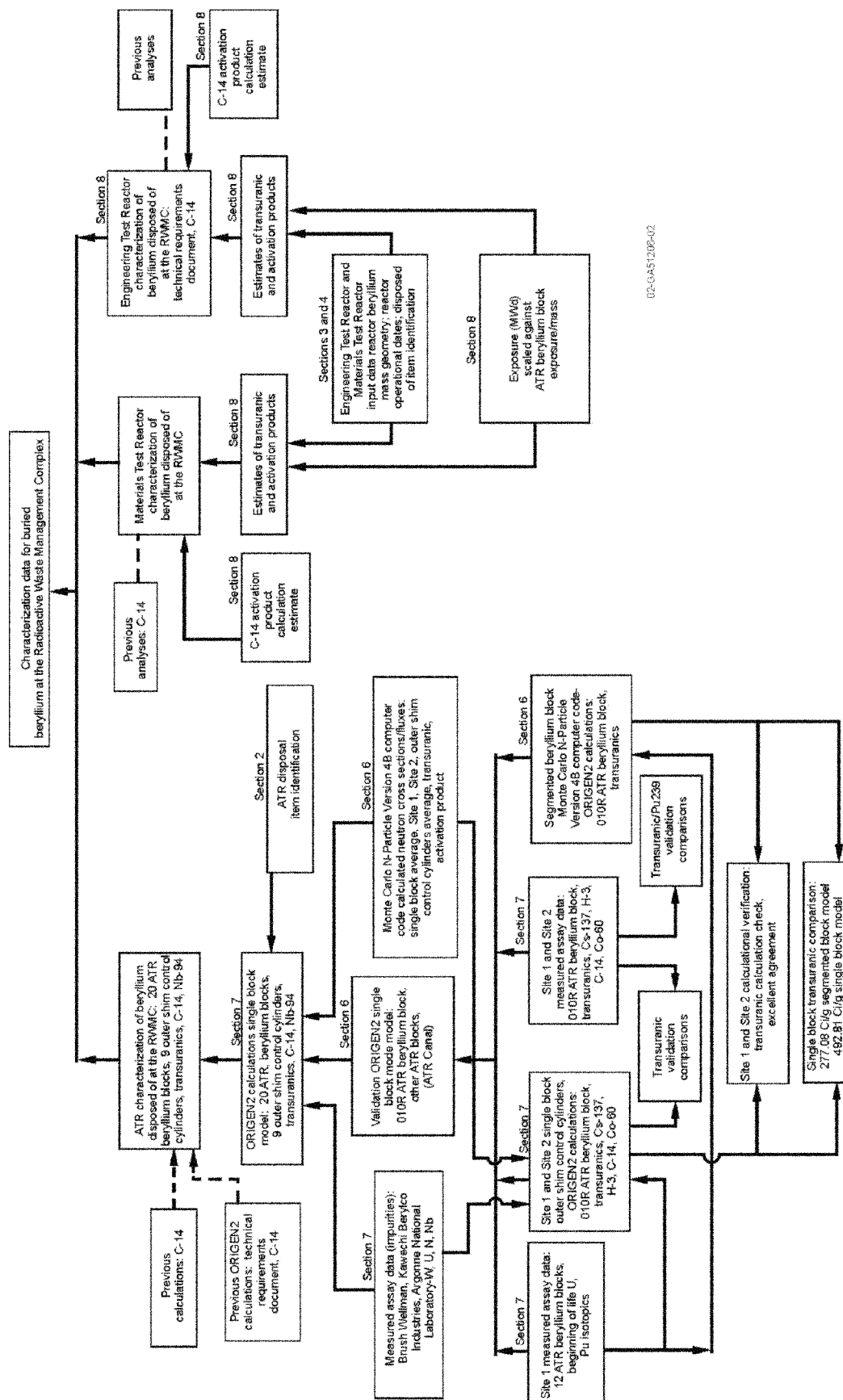


Figure 1-3. Characterization methodology for beryllium from the Advanced Test Reactor, Materials Test Reactor, and the Engineering Test Reactor buried in the Subsurface Disposal Area.

1.6 Report Organization

A summary of each section is given as follows:

- Section 1 presents the introduction, purpose, scope, background, findings, and overview of methodology and assumptions.
- Section 2 summarizes the configuration of the ATR reactor and its operational history. This section describes the beryllium reflector material used in the ATR beryllium blocks, the suppliers of the beryllium material, the inventory of the beryllium components disposed of, disposal dates, and location of items buried in the SDA.
- Section 3 summarizes the configuration of the MTR reactor and its operational history. This section describes the MTR beryllium material and identifies similarities to the ATR beryllium, the inventory of the beryllium components disposed of, disposal dates, and location of items buried in the SDA.
- Section 4 summarizes the configuration of the ETR reactor and its operational history. It describes the ETR beryllium material and similarities to the ATR beryllium, the beryllium components disposed of, disposal dates, and location of the items buried in the SDA.
- Section 5 discusses the future generation of and possible future disposal of the ATR, ETR, and MTR beryllium reflector waste as remote-handled LLW in the SDA active pit are discussed.
- Section 6 describes the calculation techniques and tools employed to accurately predict activation product concentrations or radionuclide inventories for the ATR irradiated beryllium Block 010R.
- Section 7 describes the ORIGEN2 input models used to calculate the radionuclide concentrations in the ATR beryllium reflector blocks disposed of in the SDA.
- Section 8 presents the conclusions and recommendations for characterizing the MTR, ETR, and ATR beryllium reflector material buried in the SDA.

2. ADVANCED TEST REACTOR BERYLLIUM MATERIAL

This section describes the beryllium reflector material used in the ATR reactor and its physical configuration, weight, neutron exposure, and the location in the reactor. This section also reviews waste disposal documents to support the specific identification and number of the ATR beryllium reflector material items disposed of in the SDA. Results from analysis of ATR beryllium are later used to estimate inventories in other beryllium buried at the SDA.

2.1 Reactor Reflector Description and Operational History

The ATR, like some other research reactors, has a beryllium reflector to intensify the neutron flux in the core. The ATR reflector consists of a set of eight blocks and 16 OSCCs. One such block is shown in Figure 2-1. Each block has a mass of about 81,420 g (179.5 lb) after machining. Hence, a set of eight blocks would contain 651,360 g (1,436 lb) of beryllium. Block metal volume after machining is 0.044 m³. Each block is 129.5 cm (51 in.) long (Erickson 1966). The cross-section of a block can be fit in a circle nominally 51 cm (20 in.) in diameter. The relative placement of these blocks in the ATR core is shown in Figure 2-2.

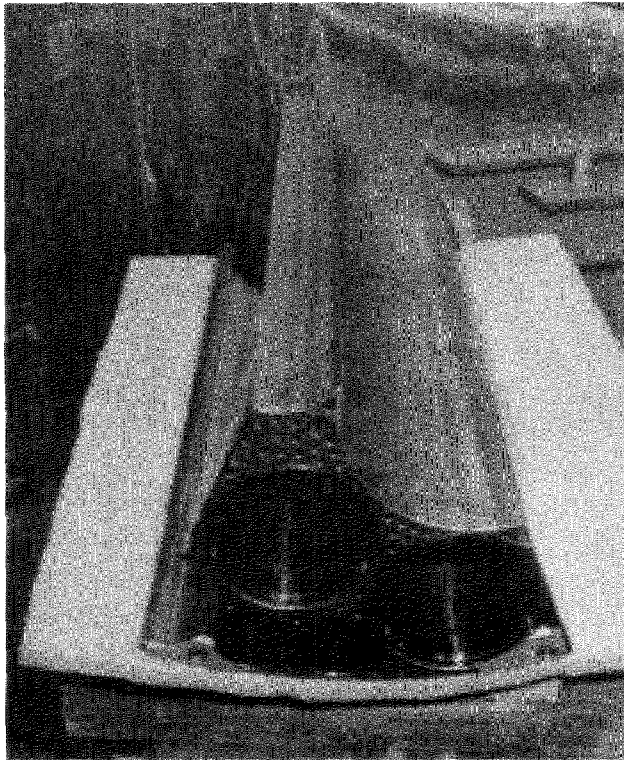


Figure 2-1. Photograph of an Advanced Test Reactor beryllium reflector block.

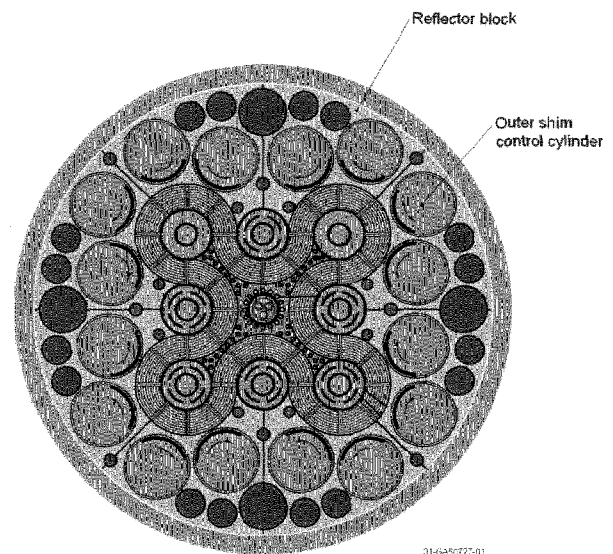


Figure 2-2. Cross-sectional view of beryllium block and outer shim control cylinder placement in the Advanced Test Reactor core.

The (n,2n) and (n, α) reactions that accompany the use of beryllium in reactors generate substantial amounts of He-4, He-3, and H-3. The accumulation of helium and hydrogen atoms causes the beryllium to swell, requiring it to be replaced periodically. Since the ATR began operation in 1967, the beryllium reflector blocks have been replaced in three planned core internal changeouts. These CICs took place in

1977, 1986, and 1994. The first CIC, in the early 1970s, was not planned though the beryllium blocks were changed out. The OSCCs, installed during original plant construction, were reused for reflector Core 2, and changed out in 1977 (see Attachment A of “Assessment of Neutron-Activation Products in Low-Level Waste Discharged from Nuclear Reactor at the Test Reactor Area and Sent to the Radioactive Waste Management Complex for Disposal.”).^a Including the original reflector, four beryllium reflectors (32 blocks) have been removed from the ATR. The fifth beryllium reflector currently remains in the ATR. A sixth reflector has been procured for the 2004 CIC.

The OSCCs are circular cylinders 18.4 cm (7.25 in.) in diameter and 119 cm (46.8 in.) long. They hold hafnium plates used to control the reactor flux. The OSCC weight in air, excluding the weight of attached hafnium components, is 57 kg (125 lb). Early OSCCs were single pieces, but later ones were built up of three shorter cylindrical sections. Each core has 16 OSCCs (48 segments). The 16 OSCCs from Core 1 were reused in Core 2. Placement of the OSCCs in the ATR core is shown in Figure 2-2.

2.2 Reflector Material and Chemical Impurities

Beryllium reflector material used in the ATR beryllium blocks has come primarily from two suppliers. Kawecki Berylco Industries (KBI) provided the reflector blocks for Cores 1 through 4. Defunct since 1979, KBI was located in Reading, Pennsylvania. Brush Wellman produced the blocks in Core 5 (currently in the reactor) and Core 6 (to be installed in 2004). All blocks disposed of in the SDA have come from Cores 1, 2, and 3.

Elemental composition of the beryllium varies from manufacturer to manufacturer depending on the source of the beryllium ore, the amount of blending of different ore sources, the amount of recycled beryllium used from unknown sources, and to some degree, the manufacturing process used to grind the beryllium particles to the correct size and press the beryllium billets. The beryllium reflector material used in the ATR OSCCs also has come from both of these suppliers. However, all of the nine OSCCs disposed of in the SDA are known to be provided by KBI (because they were from Cores 1 and 2 and were provided in the initial procurement). The specific ATR reflector material and chemical impurities used for radiological characterization modeling is fully described in Section 7.

2.3 Disposal of Advanced Test Reactor Beryllium Material in the Subsurface Disposal Area

To validate the disposal of ATR beryllium material in the SDA, the authors reviewed material in Logan (see Footnote A) and Moncur,^b the Radioactive Waste Management Information System (RWMIS) waste disposal database, individual waste shipment records (Idaho Operations Office Waste Disposal Request and Authorization Record), and the Sampling and Analysis Plan for ATR Beryllium (Haney 1999). None of the individual references provided a comprehensive or correlated inventory of ATR beryllium items disposed of in the SDA. An attempt to correlate disposal information between each of the references to develop a comprehensive inventory was not successful. Neither the RWMIS database records nor the individual disposal and shipment records provided sufficient information to clarify

a. Logan, J. A., 1999, Interdepartmental Correspondence to T. L. Clements, Jr., G. E. Ellis, and W. W. Gay, September 9, 1999, “Assessment of Neutron-Activation Products in Low-Level Waste Discharged from Nuclear Reactor at the Test Reactor Area and Sent to the Radioactive Waste Management Complex for Disposal,” JAL-04-99, Idaho National Engineering and Environmental Laboratory, Idaho Falls, Idaho.

b. Moncur, J. Blair, Interdepartmental Correspondence to C. K. Mullen, March 3, 1998, “Beryllium Blocks,” Idaho National Engineering and Environmental Laboratory, Idaho Falls, Idaho.

disposal of specific ATR beryllium materials. The RWMIS and shipment records were either incomplete or inconsistent in identifying the specific beryllium materials disposed of and the dates of disposal.

Gay (see Footnote A) used a mass balance approach to determine the beryllium blocks and OSCCs that were buried in the SDA. This approach inventoried the beryllium reflector components remaining in the ATR canal and subtracted this amount from the total activated material that was generated over the operating period of the ATR. This approach correctly identified the total number of 20 beryllium blocks and nine OSCCs by core number that were disposed of in the SDA. However, none of the references provided individual beryllium reflector serial numbers, except for the six blocks from Core 3.

In 1999, samples were collected from the 12 beryllium blocks remaining in the ATR canal to analyze for C-14 and nitrogen. This sample collection is documented in the *Abbreviated Sampling and Analysis Plan for ATR Beryllium Blocks Supports – Phase IA of the ATR Comprehensive Beryllium Disposition Plan* (Haney 1999) and associated laboratory logbook entries. Individual beryllium block serial numbers were recorded. This information allowed development of a core-specific reactor lobe position beryllium block inventory to be used for calculating the isotopic concentration characterization for each of the 12 blocks remaining in the ATR canal and the 20 blocks disposed of in the SDA. The new block serial number inventory revealed that previous information reported in Gay (see Footnote A) and used in subsequent references, was in error. It had been previously reported that eight blocks from Cores 1 and 2 and four blocks from Core 3 had been disposed of. However, distinguishing beryllium block core design characteristics (i.e., saw cuts) observed during sample collection and reported in the laboratory logbook indicated that two of the blocks remaining in the canal could have been from Core 2 only. The new information indicates six blocks (not eight blocks) from Core 2 were disposed of in 1977.

Some questions remain concerning which specific blocks were disposed of between May and June 1977, because the references do not identify the blocks by serial number (which could be used to confirm which core they were from). However, information provided in Logan (see Footnote A, Table 5) indicates that six blocks from Core 2 were disposed of between May and June 1977. The sampling and analysis plan for ATR beryllium (Haney 1999) indicates that two blocks from Core 2 remain in the ATR canal. This confirms that six blocks from Core 2 were disposed of. The reactor discharge date (April through August 1977) and disposal date (May through June 1977) correlate and support the conclusion that six blocks from Core 2 would have been available for disposal. No blocks from Cores 3 or 4 were available because they were discharged from the reactor in 1986 and 1994.

No sampling and analysis has been performed on OSCC beryllium. Using SDA disposal dates, mass balance approach, and canal operator information, the authors determined that the nine OSCCs disposed of in the SDA could have come from Cores 1 and 2 only. (Core 1 OSCCs were reused in Core 2). No known records exist and no serial numbers are recorded for those nine OSCCs.

This analysis indicates that the ATR disposals of beryllium material in the SDA were made as follows:

- November through December 1976: eight blocks from Core 1 and 0 OSCCs.
- May through June 1977: six blocks from Core 2 and 0 OSCCs.
- August through September 1987: 0 blocks and nine OSCCs from Cores 1 and 2. (Note that Cores 1 and 2 used the same OSCCs.)
- May through June 1993: six blocks from Core 3 and 0 OSCCs.

The analysis indicates a total of 20 blocks and nine OSCCs were disposed of in the SDA from 1976 through 1993.

Disposal locations in the SDA and disposal dates for the ATR beryllium blocks and OSCCs are identified and reported in Logan (see Footnote A, Table E-1).

3. MATERIAL TEST REACTOR BERYLLIUM MATERIAL

This section presents information on the beryllium reflector materials used in the MTR. The description includes the physical configuration, weight, neutron exposure, and the location in the reactor. The manufacturer of the reflector and the relationship to the ATR beryllium reflector is evaluated. The authors reviewed waste disposal documents to support the assumption that the first MTR beryllium reflector material was most likely disposed of in the SDA.

3.1 Reactor Reflector Description and Operational History

A cross section of the MTR reactor is depicted in Figure 3-1. The shaded segments represent the beryllium reflector. It comprised a large number of pieces, each 99 cm (39-3/8 in.) high. Some were in the general shape of fuel elements and were located with the fuel elements inside the core. Others were shaped prismatic blocks that fit within the spaces between the various MTR structures. Some were essentially rectangular. Others were wedges. Many had holes for test capsules. They also had a large number of flow passages for the water coolant. The estimated weight of the beryllium reflector is about 2,000 kg (4,410 lb).

The first beryllium reflector functioned in the reactor from when it first went critical (became operational) on March 31, 1952, until July 3, 1969, when the reactor was shut down to change the fuel configuration (Rolfe and Wills 1984, pp. 11 and 43).

Beryllium can be extracted from many different minerals, mined in a number of different countries, notably Kazakhstan (Russia), Brazil, Zambia, Argentina, Australia, Angola, and Rwanda. (Floyd and Lowe 1979, p. 3) In recent decades, nearly all U.S. beryllium has been manufactured from Spor Mountain bertrandite ore mined at the Brush Wellman mine in Delta, Utah. The ore from Spor Mountain is known to contain relatively high levels of uranium and gold impurities. Production from the Delta, Utah, mine did not start until 1969, 12 years after the ETR operations began and after the MTR operations had essentially finished. Therefore, the ETR and MTR beryllium could not have been produced from the Brush Wellman mine at Spor Mountain.

Beryllium, with exceptionally low uranium impurity, is known to come from the Ulba Metallurgical Plant in Kazakhstan (Russia),^c but the United States would not have been purchasing this strategic material from the Soviet Union in the early 1950s. Likewise, the United States probably would not have been buying it from China. More likely, either Brush Wellman or KBI would have furnished the ETR beryllium than either the French or Indian companies. The authors conclude that the most likely source of the MTR and ETR beryllium metal was KBI.

Before the discovery of the Spor Mountain bertrandite deposit in 1959, most of the domestic beryllium reserves were as beryl and other minerals in spodumene deposits in South Carolina (Floyd and Lowe 1979, p. 2). The MTR and ETR beryllium could have come from South Carolina, though most of the world-market beryllium in those years came from Brazil (Hanafee 1999). The ETR and MTR beryllium probably would have similar uranium impurities regardless of whether it was furnished by Brush or by KBI.

A further point is the propensity of a consumer to stay with the same source of supply for many critical components. Hence, KBI or one of its predecessors is assumed to have furnished the MTR beryllium.

c. Jacobson, L., Los Alamos National Laboratory, Personal Conversation with Glen R. Longhurst, June 2001, Idaho National Engineering and Environmental Laboratory, Idaho Falls, Idaho.

TRU, uranium is the only impurity of concern, though others, such as N-14 and Nb-93, certainly will affect the β^- emitters for determination with respect to the remedial investigation risk assessment for the OU 7-13/14 RI/FS. Concentrations in those samples were 205 ± 41.2 wppm for N-14 while concentrations were 11.7 ± 8.3 wppm for Nb-93.

3.2 Disposal of Materials Test Reactor Beryllium Waste

The authors initially analyzed existing references (see Footnote A) (Rolfe and Wills 1984) to review previous research evaluating the disposal history of the MTR beryllium. They then reviewed the INEEL RWMIS and Idaho Operations Office Waste Disposal Request and Authorization Records (disposal records) to validate the disposal information in the references and to ascertain if more detailed information may be listed on the actual disposal records. This review validated the MTR beryllium waste disposal data. Best available information indicates that the MTR beryllium disposals were made from May through June 1977 and included nine shipments and one entire reflector from Core 1.

Subsurface Disposal Area disposal locations and disposal dates for the MTR beryllium reflector material are identified and reported in Logan (see Footnote A, Table E-1).

4. ENGINEERING TEST REACTOR BERYLLIUM MATERIAL

This section describes the beryllium reflector material used in the ETR reactor. The relationship of the ETR beryllium to the ATR beryllium is developed, and the similarities are discussed. A review of waste disposal documents supports the assumption that the first ETR beryllium reflector material was most likely disposed of in the SDA.

4.1 Reactor Reflector Description and Operational History

The configuration and arrangement of the ETR beryllium reflector is shown schematically in Figure 4-1. The original ETR reflector consisted of four blocks, each $11.4 \times 88.9 \times 95.3$ cm ($4.5 \times 35 \times 37.5$ in.), that were linked together at the corners to completely surround the core laterally. The link plates were probably made of beryllium and had lifting balls on the ends to facilitate removal. Each of these blocks was penetrated by either seven or nine vertical test holes (seven on the west side, nine on the other three sides). Each block had two 4-cm (1.6-in.) diameter holes, and the rest were 3.3 cm (1.3-in.) in diameter. Each block also had a large number (98 or 99) of 6.3-mm (1/4-in.) coolant passage holes.

The first ETR reflector was removed in March 1970 and disposed of in the SDA. The second reflector was essentially the same configuration, except it was subdivided into ten vertical layers (40 pieces in all). The ETR was placed in standby status in 1980 and never restarted. The second beryllium reflector remains in the ETR today.

The mass of the beryllium from ETR Core 1 is estimated to be 624 kg (1,376 lb) from the geometry and beryllium specific gravity of 1.85. That does not include filler pieces in the test holes or the lifting balls on the link plates. Thus, each block in the first reflector would weigh about 156 kg (344 lb).

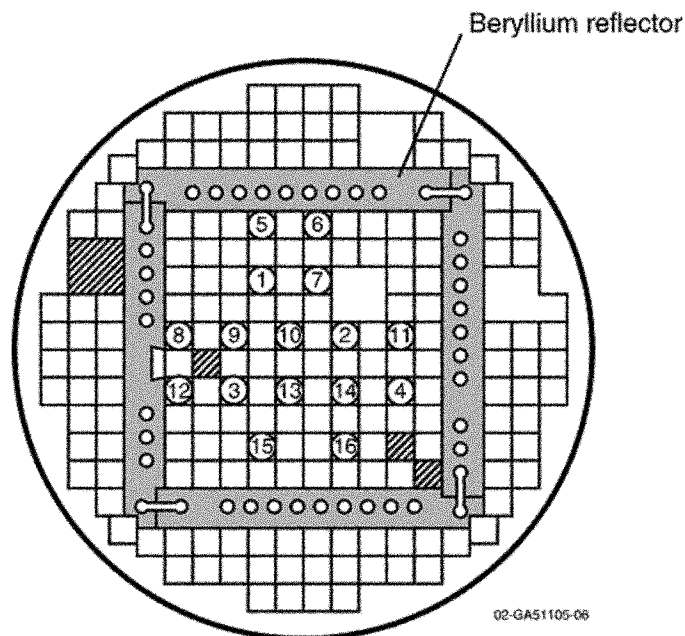


Figure 4-1. Beryllium reflector in the Engineering Test Reactor.

4.2 Reflector Material and Chemical Impurities

The initial impurity composition of the beryllium in the ETR is not known. Typical impurity levels are strongly correlated with the source of the ore from which the beryllium is reduced and less so with the manufacturing method. It is suspected that beryllium for the ETR was manufactured from the same beryllium ore used by KBI for the early ATR reflector blocks.

The discussion provided for the MTR beryllium in Section 3 also is applicable to ETR beryllium.

4.3 Disposal of Engineering Test Reactor Beryllium Waste

The authors reviewed the shipping records in the INEEL RWMIS database and the Idaho Operations Office Waste Disposal Request and Authorization Records (i.e., disposal records) that pertain to the disposal of beryllium waste. In addition, several disposal records located by Bruce Becker (i.e., TRA603SR004/21/708005015, TRA603SR004/21/7081014017, and TRA632SR010/03/731005) that pertain to ETR beryllium shipments were reviewed. All of the reviewed information indicates that most, if not all, of the beryllium waste from the first ETR core was disposed of in the SDA. In particular, the records identified by Bruce Becker indicate that approximately 64% (based on mass of material shipped) of the first ETR reflector is located in Trenches 52 and 57. However, the location of the remaining portion of this reflector is not known. The second ETR reflector remains in the ETR reactor core.

4.4 Engineering Test Reactor Critical Facility

The ETRC facility was located in TRA-654 in the southeast corner of the MTR Reactor Services Support Building (TRA-635), next to the ETR building. The ETRC was a full-scale, low-power nuclear mockup reactor of the ETR reactor. The ETRC had a beryllium reflector that was a replica of the ETR.

The ETR Characterization Report (Kaiser et al. 1982, p. 35) stated that, “. . . the [ETRC] canal was drained and left covered with plywood sheeting. . .” and that “all radioactive components have been removed from the facility.” These statements indicate that the beryllium reflector had been removed. Based on this statement, the ETRC beryllium reflector is assumed to be disposed of at the SDA. However, no reference to an ETRC beryllium reflector disposal was found in the INEEL RWMIS, other disposal records, or references investigated. Kaiser et al. (1982) issued *Characterization of the Engineering Test Reactor Facility—September 1982*; therefore the ETRC beryllium reflector is assumed to have been disposed of before that date.

The ETRC first went critical on May 20, 1957 (Burdick and Henscheid 1958, p. 4). While it was capable of 75 kW_{th}, the high-power trip point was never set above 50 kW_{th} (EG&G 1979, pp. 4–6). The average neutron flux in the fuel elements was 2.72×10^8 n/cm²/second, and the average power was only 216 W_{th} (Burdick and Parry 1958). The values markedly contrast with the high flux value in the ETR, which was 8×10^{14} n/cm²/second at the rated power of 175 MW_{th} (Kaiser et al. 1982, p. 3). Thus, the average ETRC flux was more than two million times lower than that for the ETR.

Approximately two-thirds of the beryllium used for the ETRC reflector came from modified beryllium pieces that were available from another facility. The remaining one-third of the reflector was made from sintered beryllium oxide and was contained in aluminum cans.

For the MTR reflector disposal, the authors assume that the entire irradiated beryllium reflector was disposed of in the SDA, and that none of the beryllium has been reused in the ETRC. In addition, they assume that the ETRC beryllium reflector had no preirradiation history or preradological

characteristics. The only neutron activation in the ETRC beryllium reflector was from the ETRC itself and not from prior MTR activation.

While the detailed operating history for the ETRC is not known, assuming that it operated only for the same period as the ETR would be highly conservative. The ETRC duty factor was probably only 1%. The ETR first went critical in October 1957, with full power being achieved in 1958, and continued until 1981. The ETR reflector was replaced in March 1970.

Hence, the neutron fluence in the ETRC reflector was at the most one millionth of that in the ETR reflector and probably only one hundred-millionth (10^{-8}). A simple activation model for typical beryllium from that era in an average neutron flux of 2.72×10^6 n/cm²/second gives TRU activity levels essentially just those for the inherent uranium impurity plus an equivalent level of Pa-234 that saturates after about 7 months. Increasing the neutron flux by a factor of 100, corresponding to operation at a power in excess of 20 kW_{th}, does little to change that result, only extending the time required to reach steady state.

Based on the above analysis, no issues relating to TRU are associated with the ETRC reflector, the total activity being only about 0.02 nCi/g. While not explicitly discussed here, activation of the beta emitters C-14 and Nb-94 also would be well below any threshold value of concern. They were only twice the greater-than-Class-C thresholds (10 CFR 61, 2002) at the full ETR power levels, and for them, production corresponds very closely to total neutron fluence.

